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A Concise Physical Interpretation of Several Analytical Grüneisen Formulations

by Steven B. Segletes

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Steven B. Segletes
Weapons and Materials Research Directorate, ARL

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14. ABSTRACT

A novel method is presented for deriving the Grüneisen formulations of three classical Grüneisen models. Unlike past studies which have attempted to differentiate between these Grüneisen models on the basis of Poisson ratio arguments, the current approach employs elementary vibration analysis to reach its conclusions. The current approach provides a physical insight into how the models' underlying assumptions are responsible for the analytical distinctions between the models.

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1. Introduction

The Grüneisen function, Γ, is a key thermodynamic function in the study of materials subjected to the high pressures of shock loading. Macroscopically, it relates the pressure changes to changes in internal energy at constant volume, by way of the relation

$$\Gamma = V(\partial p/\partial E)_V \quad , \tag{1}$$

where p is pressure, E is internal energy, and V is the volume of the material's unit cell. In general, Γ may be a function of the lattice volume (*i.e.*, spacing of the atomic oscillators) as well as the temperature (*i.e.*, amplitude of the vibration). However, Grüneisen theory assumes that the vibrational amplitude is small relative to the lattice spacing; thus, the set of oscillators comprising the lattice possess frequencies that are assumed to vary with volume alone. In terms of this characteristic vibrational frequency of the lattice, ω, the volume dependence of the Grüneisen function is expressed, according to classical theory (*1*, *2*), as

$$\Gamma/V = -\frac{d\omega/dV}{\omega} \quad . \tag{2}$$

During the decades in which the classical approach to lattice vibration held sway, a number of analytical Grüneisen formulations were presented in the literature, with several gaining a widely cited reputation (*1*, *3*, *4*). Because of the manner in which these particular models were formulated, Γ was given in terms of both the material's volume and cold pressure, p_c (*i.e.*, the $0°$ isothermal pressure). However, since the cold pressure is, itself, a function of volume alone, these formulations for Γ indeed satisfy the Grüneisen assumption of temperature independence.

While these important models retained the key definitional coupling embodied in equation 2, many later developers of numerical hydrocodes for impact simulation opted to bypass the formalism of these historical models, in favor of direct empirical characterization of $\Gamma(V)$ according to preferred functional forms. Perhaps the motivation in this was that empirical shock data provided a material's shock Hugoniot, and not the cold-pressure curve demanded of the classical models. Regardless of the motivation, once analytically decoupled from the mechanics of lattice compression, these wholly empirical formulations for Γ were not always thermodynamically robust. Segletes (*5–7*) has shown how these decoupled Grüneisen formulations are susceptible to an unusual host of thermodynamic inconsistencies that can lead to nonphysical thermodynamic instabilities in shock/impact calculations.

There is value, then, in reconsidering the classical Grüneisen methods as an important bridge between modern *ab initio* methods and the decoupled empirical approaches that still characterize many extensively utilized numerical codes in the shock/impact community.

2. Classical Grüneisen Theories

The classical models by Slater (*1*), Dugdale and MacDonald (*3*), as well as the free-volume theory cited by Vashchenko and Zubarev (*4*), have been widely studied and utilized and relate a materials' Grüneisen behavior to the compression behavior of the lattice. While it is somewhat difficult to compare the original derivations of these three models side by side to ascertain the particular points of departure that bring about their distinctions, some have attempted to do exactly this.

While the forms originally presented by Slater,

$$\Gamma = -\frac{2}{3} - \frac{V}{2}\left(\frac{d^2p_c/dV^2}{dp_c/dV}\right) \quad , \tag{3}$$

and that of Dugdale and MacDonald,

$$\Gamma = -1 - \frac{V}{2}\left(\frac{d^2p_c/dV^2 - 10p_c/9V^2}{dp_c/dV + 2p_c/3V}\right) \quad , \tag{4}$$

bear a notable similarity as presented, it was Vashchenko and Zubarev (*4*) who were able to present a unified view of these results, along with the free-volume theory. They present a single parameterized algebraic expression which accurately captures the forms of all three of these models, depending upon the value of the model's parameter. This expression is given as

$$\Gamma = -\frac{4-3n}{6} - \frac{V}{2}\frac{\dfrac{d^2}{dV^2}(p_cV^n)}{\dfrac{d}{dV}(p_cV^n)} \quad , \tag{5}$$

where n is the model parameter. When n takes on values of 0, 2/3, and 4/3 respectively, the models of Slater (*1*), Dugdale and MacDonald (*3*), and the free-volume theory (*4*) are respectively recovered.

To arrive at equation 5, Vashchenko and Zubarev postulated a hypothetical form for the Poisson ratio, ν, in terms of the cold pressure, volume, and model parameter n:

$$\nu = \frac{1 - \dfrac{1-\nu_0}{1+\nu_0}\left(1 + n\dfrac{d\ln V}{d\ln p_c}\right)}{1 + \dfrac{1-\nu_0}{1+\nu_0}\left(1 + n\dfrac{d\ln V}{d\ln p_c}\right)} \quad . \tag{6}$$

Coupled with certain relations specifying the manner in which the Poisson ratio relates to the lattice vibrational frequencies, equation 6 was shown to lead directly to equation 5.

Therefore, if one accepts their postulated form for the Poisson ratio, equation 6, then the three Grüneisen models of interest (*1, 3, 4*) follow from equation 5 as specific instances of the proposed form. Pastine (*8*) has also utilized an explanation involving the influence of the Poisson ratio as a means to interpret and differentiate expressions for the Grüneisen formulation.

Conceptually, such an approach makes sense, and there is certainly nothing invalid about it. However, the intuitive understanding that one may obtain from equation 6 is, at best, quite limited, especially when the Poisson form is, itself, expressed in terms of volume derivatives of the cold pressure.

What is more, the original derivations of the models of Slater, Dugdale and MacDonald, and the free-volume theory were not made on the basis of this or any other Poisson ratio formulation. That researchers could, after the fact, discover the Poisson ratio as a means to differentiate the models was a useful and convenient resolution to the useful question of "what scientific explanation can explain the functional difference between these three models?" But, because the original models were never derived in terms of Poisson ratio, the Poisson explanation fails to answer the more fundamental question of "what is it about the formulation of these three models that leads to their functional differences?"

3. Result

In this report, a different approach is taken to compare these historical models, based upon treating the material lattice as a spring-like structure, in which the various models may be differentiated on the basis of what kind of spring composes the lattice and what measure is taken to quantify the spring's characteristic spatial dimension. The key link in eventually relating the spring's response back to the Grüneisen function is the classical equation 2, which defines the Grüneisen function in terms of the characteristic vibrational frequency of the lattice ω.

Consider a lattice with spacing λ such that $\lambda^3 = V$. When atomic vibration is not considered (*i.e.*, at $0°$ temperature), the potential energy of the lattice network is defined by the cold energy of the lattice E_c, which will be a function of only a spatial dimension that is characteristic of the lattice, call it ξ. This characteristic dimension might take on units of length (as in the lattice spacing λ), though it need not necessarily do so; rather, it could take the form of volume V or some other spatially related quantity.

For a quantity characterized by an isotropic field potential, such as the lattice energy E_c, the negative of the gradient of the potential with respect to ξ, in this case $-\nabla E_c(\xi)$, is an indicative measure of the field's "force," while the Laplacian of E_c, that is $\nabla^2 E_c(\xi)$, is a measure of the field's "stiffness." The terms "force" and "stiffness" are quoted here because depending on the nature of the characteristic dimension ξ, the physical units of the characteristic lattice "force" might be other than force and likewise for the "stiffness."

Vibration analysis indicates that the system's characteristic stiffness is associated with a characteristic vibrational frequency ω_ξ according to the following relation:

$$\nabla^2 E_c(\xi) \propto \omega_\xi^2 \quad , \tag{7}$$

where the symbol "$\propto$" denotes a direct proportionality. This expression has been used in the study of lattice mechanics (*9, 10*). As in the case of the characteristic force and stiffness, the ξ-based characteristic frequency ω_ξ may not have the physical units of 1/time that are traditionally associated with frequency, though it will have dimensions of propagation speed per unit characteristic dimension. However, it may be related to a characteristic frequency ω that does have traditional units (*i.e.*, 1/time). This connection is made through the definitional relations of vibration theory which connect the characteristic frequency and propagation velocity through the vibrational wavelength:

$$\omega_\xi \xi = C = \omega\lambda \quad . \tag{8}$$

Thus, combining equations 7 and 8 gives the following proportion:

$$\nabla^2 E_c(\xi) \propto \left(\frac{\omega\lambda}{\xi}\right)^2 \quad . \tag{9}$$

Equation 9 is the simple relation from which the results of this report flow. Applying it to the task of acquiring a relationship for the Grüneisen function is accomplished by starting with equation 9 and isolating ω^2. Take the derivative of this equation with respect to volume V and divide the resulting equation by the original equation. The result, in light of equation 2, is given as

$$\Gamma = -\frac{V}{2}\,\frac{\dfrac{d}{dV}\left(\dfrac{\xi^2}{\lambda^2}\nabla^2 E_c(\xi)\right)}{\left(\dfrac{\xi^2}{\lambda^2}\nabla^2 E_c(\xi)\right)} \quad . \tag{10}$$

There are really two parameters that characterize equation 10. The first is the characteristic lattice dimension ξ, and the second is physical nature of the lattice spring whose stiffness is characterized by $\nabla^2 E_c(\xi)$. Depending on whether the lattice spring is considered a simple one-dimensional (1–D) spring or a three dimensional (3–D) radial spring, the Laplacian operator would be expressed as

$$\nabla^2 E_c(\xi) \propto \begin{cases} d^2E_c/d\xi^2 & \text{(simple 1–D spring)} \\ d^2E_c/d\xi^2 + (2/\xi)dE_c/d\xi & \text{(3–D radial spring)} \end{cases} \quad . \tag{11}$$

From equation 10, in light of equation 11, it may be directly shown that Slater's relation is recovered when the lattice spring is a simple 1–D spring and the characteristic spatial dimension ξ of the lattice is taken as its cell volume V. Similarly, the relation of Dugdale and MacDonald is recovered by taking the lattice spring to be simple 1–D, while taking the characteristic spatial dimension ξ as the lattice spacing λ. Finally, the free-volume theory may be recovered* from equation 10 when the lattice spring is taken as a 3–D radial spring and the characteristic spatial dimension ξ of the lattice is the lattice spacing λ. Table 1 categorizes some of the relevant quantities for these three models.

Table 1. Characteristic spring type and spatial dimension for various Grüneisen theories and physical units of associated characteristic quantities.

Theory	Lattice Spring Type	Characteristic Spatial Dimension, ξ	Units of Characteristic	
			Force	**Stiffness**
Slater	Simple (1–D)	V	Pressure	Pressure/volume
Dugdale-MacDonald	Simple (1–D)	λ	Force	Force/length
Free-volume theory	Radial (3–D)	λ	Force	Force/length

These differentiating characteristics are not merely coincidental, which just happen to produce the given models; rather, they are indicative of the logic and methodologies employed by the researchers in their original derivations. In the case of Slater's derivation, a constant Poisson ratio was assumed. When this is stipulated for a material, the elastic and bulk moduli remain proportional. This allowed the vibrational frequency to be cast in terms of volume derivatives of pressure. Thus, it is not surprising that the characteristic dimension of the Slater model is volume V, leading to a stiffness expressed in terms of pressure per volume.

In the case of the Dugdale-MacDonald model, the authors' explicit contention was that the characteristic lattice velocity is always of a form consistent with a 1–D spring formulation (which is to say, with a characteristic stiffness having units of force per length). Plendl (*12*) has also asserted this relationship. Dugdale and MacDonald even went so far as to show that Slater's model failed to give the proper Grüneisen value of zero for a harmonic lattice of simple 1–D springs. Thus, wholly consistent with the model's underlying principal, a lattice of simply-connected 1–D springs would necessarily retain λ as the characteristic dimension.

The free-volume theory differs from the Dugdale-MacDonald relation only in that the lattice spring is taken as a 3–D radial spring rather than as a simple 1–D spring, reflecting a more complex view of reality in terms of the interatomic potential as a 3–D field and not as a 1–D spring.

*Recognition of the compatibility of the free-volume theory with the underlying tenets of equations 2 and 7 was also noted in unpublished work by Shanker *et al.* (*11*).

It is important to note that when ξ defines an orientation-independent lattice measure such as λ or V, equation 7 describes a volumetric oscillation, since an ordinary derivative of E_c, with respect to the characteristic dimension ξ, implies that ξ is changed between every atom in the lattice network (and in every component direction), in order to evaluate the resultant change in E_c. Therefore, in such cases, the quantity ω_ξ may be justifiably termed a "volumetric frequency," regardless of whether the springs are characterized as 1–D or 3–D.

However, it has been shown (*13*) that when the lattice interactions are restricted to nearest neighbors only (or alternately when the lattice component stiffness is purely harmonic in nature), the stiffness associated with a volumetric change is in proportion to the stiffness associated with a 1–D vibrational wave pulse. Therefore, under the assumption of nearest-neighbor interactions, this volumetric frequency ω_ξ is equally indicative of a traveling-wave vibrational frequency from which the Γ function actually arises.

In all the models examined in this report (*1, 3, 4*), the original authors explicitly invoked nearest-neighbor arguments in their derivations. This widely used nearest-neighbor assumption (*1, 3, 4, 14, 15*) serves to justify the use of ω_ξ in equation 8 as a means to link the vibrational frequencies of volumetric change to those of traveling waves. It must be remembered, however, that in the general case when distant interactions of non-nearest neighbors are considered, the volumetric frequency and vibrational frequency are not identical (*13*). As such, equation 7 remains merely an approximation when applied to the global (as opposed to the pairwise) potential.

4. Conclusion

The simple direct formulation of the Grüneisen function that herein arises from elementary vibration analysis serves to provide a clear foundational understanding of three popular Grüneisen models (*1, 3, 4*) and their differentiating characteristics. Unlike the historical explanations (*4, 8*) that, after the fact, invoked the functional behavior of the Poisson ratio as the means to functionally differentiate the historical models, the current approach offers an explanation in terms of two fundamental modeling assumptions—the nature of the interatomic "spring" and the characteristic spatial measure of the lattice. Such a differentiation, by its nature, provides a satisfactory answer to the question of “what is it about the formulation of these three models that leads to their functional differences?” Such understanding is absent with the historical explanations, where the differentiating feature is cast in terms of the Poisson ratio.

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